

## Characterization of highly crystalline C<sub>60</sub> thin films and their field-effect mobility

Th. Birendra Singh<sup>\*1</sup>, H. Yang<sup>2</sup>, B. Plochberger<sup>3</sup>, L. Yang<sup>4</sup>, H. Sitter<sup>3</sup>, H. Neugebauer<sup>1</sup>, and N. S. Sariciftci<sup>1</sup>

<sup>1</sup> Linz Institute of Organic Solar Cells (LIOS), Physical Chemistry, Johannes Kepler University Linz, 4040 Linz, Austria

<sup>2</sup> Rensselaer Nanotechnology Center, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

<sup>3</sup> Institute of Semiconductors and Solid State Physics, Johannes Kepler University Linz, 4040 Linz, Austria

<sup>4</sup> National Synchrotron Light Source, Brookhaven National Laboratory, Upton, NY 11973, USA

Received 14 May 2007, revised 29 August 2007, accepted 20 September 2007

Published online 8 November 2007

PACS 72.80.Le, 73.61.Ph, 85.30.Tv

Transistors fabricated from C<sub>60</sub> films grown by hot wall epitaxy at elevated substrate temperature of 250 °C, show charge carrier mobility of ~6 cm<sup>2</sup>/Vs. When grown at substrate temperature of 25 °C, mobilities of only 0.6–1 cm<sup>2</sup>/Vs are obtained. The C<sub>60</sub> films were characterized using grazing-incidence X-ray diffraction and show increased crystalline properties when grown at elevated substrate temperature. The improvement in the charge carrier mobility is attributed to the higher crystalline nature of the thin films.

© 2007 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

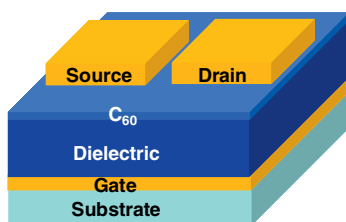
### 1 Introduction

Growth control of organic thin films is a prerequisite for the fabrication of high performance electronic devices such as organic field-effect transistors (OFETs) and logic circuits based upon them, since charge carrier mobility strongly depends on the structural characteristics of the organic films [1–3]. Recently, organic film transistors with charge carrier mobilities comparable to those obtained with amorphous silicon based devices have been demonstrated [4–6]. As a consequence, several previously envisioned applications incorporating organic materials are now becoming feasible [7–10]. C<sub>60</sub> with a large electron affinity of 2.65 eV [11] has been shown to give *n*-channel transistors with high electron mobilities [5]. The crystalline structure and the morphology of the organic semiconductor thin film can be heavily influenced by preparation conditions, e.g. the rate and the temperature (*T*<sub>s</sub>) at which the film is deposited. In this article, we report that C<sub>60</sub> thin films when grown at higher substrate temperature result in higher charge carrier mobility.

### 2 Experimental procedure

Discrete top-contact transistors (see Fig. 1) were fabricated on quartz glass substrates with a predefined ITO electrode acting as the gate terminal. The precursor compound of divinyltetramethyldisiloxane-bis(benzocyclobutene) (BCB), purchased from Dow Chemicals, was spin coated on top of the ITO/quartz

\* Corresponding author: e-mail: birendra.singh@jku.at

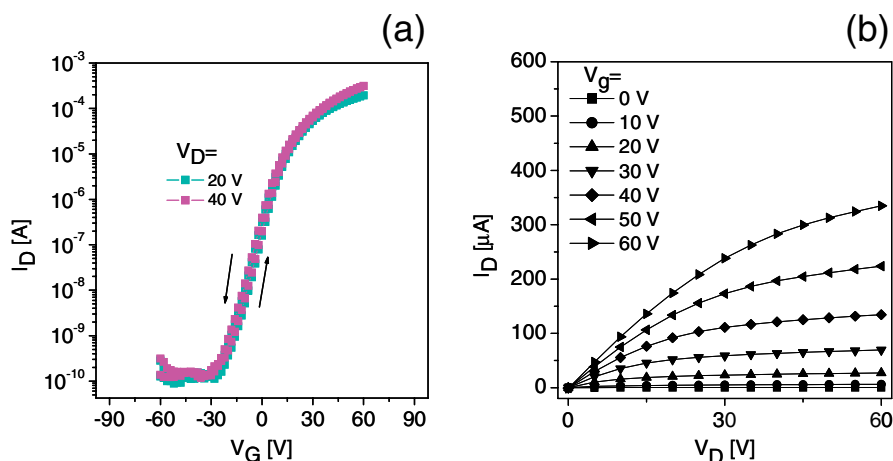


**Fig. 1** (online colour at: www.pss-b.com) Schematic structure of the organic field-effect transistor.

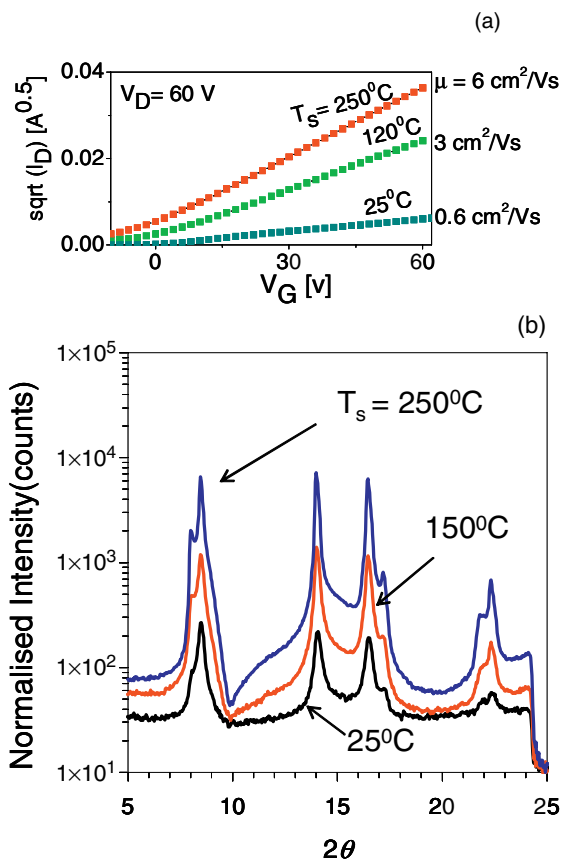
substrate. The substrate was cured at 250 °C for 2 hours in a vacuum oven, forming the BCB dielectric. Following the curing, a 300 nm thick film of C<sub>60</sub> was grown by hot wall epitaxy (HWE, [12]) at various substrate temperatures in the range from 25 °C to 250 °C. Finally, top-contact electrodes consisting of LiF/Al (0.6 nm and 60 nm, respectively) were evaporated under vacuum (~10<sup>-6</sup> mbar) through a shadow mask. The channel length and width of the transistor was 30 μm and 1.5 mm, respectively. For the X-ray characterization of the C<sub>60</sub> film, Si wafers with native oxide layers on top were used as substrates. The same growth parameters as for transistors were used to obtain similar C<sub>60</sub> films. Electrical measurements were performed at room temperature using an Agilent E5273A source-measure unit. The crystallinity of the C<sub>60</sub> films was characterized by two dimensional grazing-incidence X-ray diffraction (GIXD) measurements at the beam line X21 of the National Synchrotron Light Source at the Brookhaven National Laboratory. The sample was mounted on a two-axis goniometer on top of an *x*-*z* stage and the scattered intensity was recorded by a 2-D Mar CCD detector. The incident-beam angle was set to be 0.3° in order to increase the scattering intensity.

### 3 Results and discussion

A transistor device with C<sub>60</sub> film, which was grown at  $T_s = 120$  °C was characterized under inert conditions inside a glove box. Typical *n*-channel OFET transfer characteristics with no hysteresis behaviour were obtained, as shown in Fig. 2a. With depleted drain current  $I_D = 100$  pA at drain voltage  $V_D = 40$  V and at gate voltage  $V_G = -30$  V, and saturated  $I_D = 0.3$  mA at  $V_G = 60$  V, a saturated mobility of 3 cm<sup>2</sup>/Vs with an on-off ratio higher than 10<sup>6</sup> was obtained. The sub-threshold slope was 7 V/decade. When characterized in the linear regime at  $V_D = 20$  V, a linear mobility of 1.2 cm<sup>2</sup>/Vs was obtained. Typical output characteristics, shown in Fig. 2b, also demonstrate an *n*-channel OFET.



**Fig. 2** (online colour at: www.pss-b.com) Typical transfer (a) and output characteristics (b) of OFETs with C<sub>60</sub> films grown at 120 °C.



**Fig. 3** (online colour at: [www.pss-b.com](http://www.pss-b.com)) (a) Plot of  $\sqrt{I_D}$  vs.  $V_G$  for OFETs with  $C_{60}$  films grown at different temperatures. (b) Circular average X-ray profiles for 100 nm thick  $C_{60}$  films deposited on  $SiO_2/Si$  substrates kept at different growth temperatures  $T_s$ .

The performance of the OFET depends strongly on the substrate temperature. A plot of  $\sqrt{I_D}$  vs.  $V_G$  for OFETs with thin films grown at various substrate temperatures is shown in Fig. 3a. The results show an increase in mobility from 0.6 cm<sup>2</sup>/Vs to 6 cm<sup>2</sup>/Vs when changing the substrate temperature from 25 °C to 250 °C. To correlate the mobility with the substrate temperature during film formation we performed GIXD studies on films prepared at different temperatures. Here we used  $SiO_2$  instead of BCB to avoid large amorphous background scattering of BCB in the X-ray diffraction spectrum. A plot of  $2\theta$  scans is shown in Fig. 3b. The results show that  $C_{60}$  films grown at higher substrate temperatures have higher peak intensities compared to films grown at lower temperature, indicating that higher substrate temperature leads to higher crystallinity.

Studies of the surface morphology [13] using atomic force microscopy reveal increased crystallite sizes upon increasing the substrate temperature. The increased crystallite sizes are supposed to lead to reduced grain boundaries and to improve the charge transport properties. It is also possible that higher substrate temperature eliminates various impurities on the substrate leading to the formation of better quality  $C_{60}$  films.

#### 4 Conclusion

$C_{60}$  thin films which are grown at various substrate temperatures are characterized using GIXD. The results are compared to the measured charge carrier mobility using OFET geometry. Higher crystallinity of the  $C_{60}$  films is obtained when depositing the  $C_{60}$  films at higher substrate temperatures. A correlation

of the charge carrier mobility and the crystalline properties of the thin-films is demonstrated. The increased mobility is expected to be due to reduced grain boundaries.

**Acknowledgements** The authors acknowledge financial support from Austrian National Science Foundation, FWF project NFN No. P16891-N08 and S9706-N08. HY acknowledges financial support from the Nanoscale Science and Engineering Institute of the National Science Foundation (DMR 0117792).

## References

- [1] H. Sirringhaus, P. J. Brown, R. H. Friend, M. M. Nielsen, K. Bechgaard, B. M. W. Langeveld-Voss, A. J. H. Spiering, R. A. J. Janssen, E. W. Meijer, P. Herwig, and D. M. de Leeuw, *Nature* **401**, 685 (1999).
- [2] C. D. Dimitrakopoulos and P. R. L. Malenfant, *Adv. Mater.* **14**, 99 (2002).
- [3] R. J. Kline, M. D. McGehee, and M. F. Toney, *Nature Mater.* **5**, 222 (2006).
- [4] I. McCulloch, M. Heeney, C. Bailey, K. Genevicius, I. MacDonald, M. Shkunov, D. Sparrowe, S. Tierney, R. Wagner, W. Zhang, M. L. Chabinyc, R. J. Kline, M. D. McGehee, and M. F. Toney, *Nature Mater.* **5**, 328 (2006).
- [5] Th. B. Singh, N. Marjanovic, G. J. Matt, S. Gunes, N. S. Sariciftci, A. M. Ramil, A. Andreev, H. Sitter, R. Schwödiauer, and S. Bauer, *Org. Electron.* **6**, 105 (2005).
- [6] M. Heeney, C. Bailey, K. Genevicius, M. Shkunov, D. Sparrowe, S. Tierney, and I. McCulloch, *J. Am. Chem. Soc.* **127**, 1078 (2005).
- [7] G. H. Gelinck, H. E. A. Huitema, E. van Veenendaal, E. Cantatore, L. Schrijnemakers, J. B. P. H. van der Putten, T. C. T. Geuns, M. Beenhakkers, J. B. Giesbers, B.-H. Huisman, E. J. Meijer, E. M. Benito, F. J. Touwslager, A. W. Marsman, B. J. E. van Rens, and D. M. de Leeuw, *Nature Mater.* **3**, 106 (2004).
- [8] D. M. de Leeuw, US Patent WO99/30432 (1999).
- [9] S. Steudel, K. Myny, V. Arkhipov, C. Deibel, S. de Vusser, J. Genoe, and P. Heremans, *Nature Mater.* **4**, 597 (2005).
- [10] L. Zhou, A. Wanga, S.-C. Wu, J. Sun, S. Park, and T. N. Jackson, *Appl. Phys. Lett.* **88**, 083502 (2005).
- [11] D. L. Lichtenberger, K. W. Nebesney, C. D. Ray, D. R. Huffman, and L. D. Lamb, *Chem. Phys. Lett.* **176**, 203 (1991).
- [12] H. Sitter, A. Andreev, G. Matt, and N. S. Sariciftci, *Mol. Cryst. Liq. Cryst.* **385**, 51 (2002).
- [13] H. Yang et al., to be published.